Photonic transport: the key to the confinement of random laser modes

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A remarkable result in the research on random lasers has been the experimental finding that spatially confined and extended laser modes can actually co-exist in the same region of strongly scattering nanocrystalline powders[1,2]. This has faced theoreticians with a serious challenge[4], since an 'ab initio' description for coherent emitting modes in diffusive systems[3–7] could not yet be given for this phenomenon. In this paper we derive by means of field diagrammatical photon transport[8–12] incorporating several loss channels the co-existence of spatially confined and extended random laser modes. This theory proofs that the experimentally observed mode types in different gain regimes can be explained in a single framework, and that they are determined by loss. The diversity of loss can be caused by the particular arrangement of the scatterers, the spatial distribution of the nonlinear gain or the absorption by an adjacent substrate.

Up to the present random lasers have been one of the most controversially discussed systems in photonics[13–20]. Although statements like 'Random lasers work without an external feedback mechanism' have been stressed a good deal, the full consequences of this aspect have never been brought up. The rich physics and possible applications[21,22] behind what at first glance appears to be a simplistic system of just a 'dust powder' have barely started to reveal their secrets. Absolutely essential for the fundamental behavior of a random laser is the spatial extension of random lasing modes. If the lasing spots are small, the random laser actually is operated as a collection of single-mode lasers where the modes do not overlap in space[23]. On the other hand, the random laser can exhibit another type of mode which covers the whole ensemble showing significantly different, speckle-like emission characteristics and a higher laser threshold. The extended and confined modes may overlap in space but for that case they are spectrally well separated. Both lasing regimes have found their supporters in theory but only recently has the same ansatz been suggested as an explanation for both phenomena[4].

The scenario investigated here is random lasing in nanocrystalline ZnO (see supplement[1]) and can be described as follows: For the excitation intensity meeting the laser threshold, a spatially confined mode starts to lase and with increasing pump strength, a rising number of modes of this type may be found in a regime of weak localization. This behavior can be exactly derived by the semiclassical, field theoretical ansatz of Vollhardt and Wölfle diagramatics for light in a diffusive system including interferences. The ZnO grains are considered to be Mie spheres[24,25] with a diameter of 260 nm and the loss of the random laser sample is assumed to be unidirectional. We have implemented the permittivity using $\mathrm{Re}\{\epsilon_s\} = 4.0164$ and a self-consistently calculated imaginary part, the microscopic gain.

Increasing the excitation power in the experiment causes a striking change in the system's behavior at a certain point. Besides the small spatially confined modes, spectrally well-separated modes are observed which exhibit a completely different behavior in space (see Fig. 1). Their diameter is large compared to the others, they can cover the whole sample. So in theory that behavior can not be explained just by the increase of the pump power alone. Only investigating the possibility of loss in more than one direction, as it may be caused e.g. by the variance of the powder's granularity or by the loss due to the absorbing substrate, contributes to a better understanding. The additional loss initially leads to a suppression of a large number of modes at first which only arrive at their lasing threshold for significantly higher pump strengths when their intrinsic nonlinear gain leads to a balanced process (see Fig. 2). Strictly speaking, several distinctive loss processes yield the co-existence of both types of modes; the extended modes just lose more energy due to a higher number of loss channels than the confined modes do. So no contradiction whatsoever exists between the two regimes. The principle itself is basically reprising at another intensity scale which is induced by further degrees of freedom, namely several loss channels. One could even imagine tuning the powder's parameters in such a way that a stepwise access to differnt loss mechanisms could be possible and the random laser therefore could be controlled by the ensemble size, the surface the type of substrate etc.

What at first glance seems to be a matter of course is actually being derived by a doubly nested self-consistent consideration of the photon density response, the four-point correlator Φ, which is well known from electronics to result from the Bethe-Salpeter equation (BS) and including the coupling to a four-level laser rate equation system in stationary state. The intrinsic two-particle nature of the BS makes the calculations somehow cumbersome, since it has been shown that not only parallel propagating light paths, the ladder diagrams, contribute[12]. Especially counter propagation paths, maximally crossed diagrams called Cooperons, which resemble the propagation processes of particle and hole in electronics, lead to the description of second-order coherent emission in disordered random media.

Right here we have to turn our attention towards the dissipative processes with regard to the Green's functions formalism. It is well known that Green's functions, intended to describe the coherent intensity in the random laser system with diffusive character on the one hand but being a description of microcanonical ensembles on the

other hand, have to obey time reversal invariance. That seems to be a contradiction at first sight. However in 1931 Lars Onsager discovered that entropy was increased by processes which definitely obeyed time reversibility for their evolution[26]. In other words the damped harmonic oscillator, which can be inverted beyond doubt with respect to time, arrives at the same result in the end; thus the diffusion of light is still diffusive although light paths as such can be time reversed. As a matter of fact this simple but very helpful aspect of dissipation and disorder guarantees the feasibility of the description of propagating light intensity by the four-point correlator Φ.

The considerable effort, though, is requited beyond expectations by the highly versatile results of the multifarious self-consistent theory. Not only can the experimental findings for the light diffusion processes in random media (see Ref.[28]) be derived self-consistently, starting with the renormalized scattering mean free path l_s, leading to the full description of light diffusion including all interference effects. Also the modal behavior, the core of the random laser, is described efficiently by the determination of the coherence length ξ with respect to various loss channels. And finally the co-existence of small confined and extended modes can be completely explained.

We solve BS in a sophisticated regime of real space and momentum with respect to the dimensionality of the random laser sample and arrive at the description for the energy density $\Phi_{\epsilon\epsilon}(Q,\Omega)$, which equals the zeroth moment of the expansion of the full correlator Φ[10,12](see supplement)

$$\Phi_{\epsilon\epsilon}(Q,\Omega) = \frac{N_\omega(Y)}{\Omega + iDQ_x^2 + iD\xi^{-2}}$$

Here N_ω is the numerator basically reflecting the density of photonic modes which is responsive to the interaction with the scatterers, Q is the center of mass momentum of the propagator denoted in Wigner coordinates, Ω is the center of mass frequency and D is the self-consistently derived diffusion constant. The pump intensity is being absorbed in the vicinity of the surface, so the transport can be considered to be almost exclusively perpendicular to the direction of injection. Consequentially, loss channels occur in-plane and in the backscattering direction, which are reflected by separate dissipative length scales χ_d due to gain and absorption in-plane and ζ due to loss out of plane. The full dissipative behavior is described within a renormalized so called mass term of the diffusion equation:

$$iD\xi^{-2} = -iD\chi_d^{-2} - c_1\left(\partial_Y^2\Phi_{\epsilon\epsilon}(Q,\Omega)\right) + c_2 + iD\zeta^{-2}.$$

So it can easily be seen that modes which suffer energy loss exclusively in the backscattering direction are only influenced by a dissipative length scale which strongly depends on the self-consistent diffusion of the system. Modes losing energy to different loss channels obey a dissipative length scale which carries an inherent differential, because their mesoscopic transport character is further influenced by these losses. The coefficients c_1 and c_2 are thereby selfconsistently derived. The diffusion equation then has to be solved with respect to this distinction:

$$-\frac{\partial^2}{\partial Y^2}\Phi_{\epsilon\epsilon} = \frac{1}{D}\left[\frac{D}{-\chi_d^2} + \frac{D}{\zeta^2}\right]\Phi_{\epsilon\epsilon} + \text{spontaneous emission.}$$

The self-consistent stimulated amplification rates, in other words the nonlinear self-consistent gain $\gamma_{21}n_2$ (see supplement), carries the influences of both length scales χ_d and ζ,

$$\frac{D}{-\chi_d^2} + \frac{D}{\zeta^2} = \gamma_{21}n_2,$$

and therefore reflects the physical properties of the highly absorptive thin random laser samples, which are amongst the most strongly scattering random lasers ever investigated. The confined mode (Fig. 2) features only loss perpendicular to the surface and is therefore circularly shaped. Contrarily, the extended mode can experience in-plane losses due to the limited sample size in y-direction. These distinctive loss characteristics are responsible for the elliptical shape of the mode and its size.

We solved the system also for time-dependent laser rate[29,30] equations and derived the typical threshold behavior. The coupling of various channels of loss such as nonradiative decays enables us not only to perceive the modal regime in space but also reproduces the experimentally found threshold behavior for both regimes of lasing modes, confined as well as extended modes (see Fig. 3). The extended mode's threshold is significantly higher than the one for the confined mode.

The solution of a complicated statistical behavior like that of the random laser visualizes the power of diagrammatic transport theory which can be solved by comparably fast algorithms with respect to the length scales of diffusive propagation of light and light intensity in disordered random media. Designated 'semi-analytical calculation, microscopic theory of random lasing and light transport in amplifying disordered media proves to be a powerful tool regarding second-order coherent emission of random lasers.

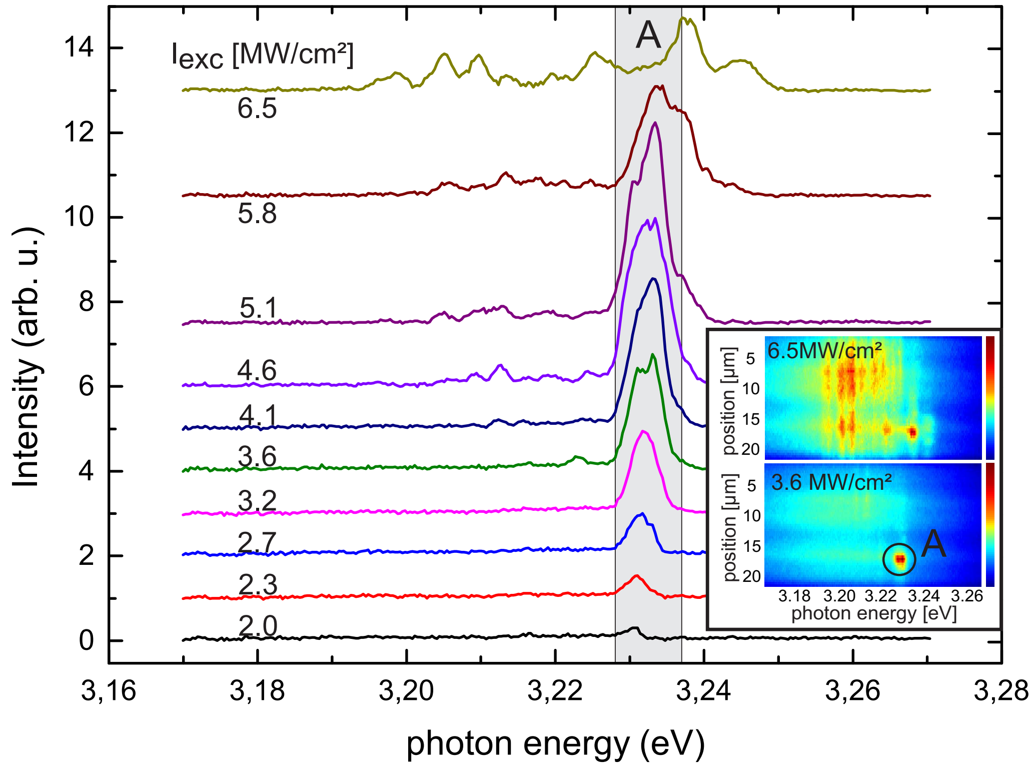

FIG. 1. Integrated spectra of a small ensemble (20x20μm) of ZnO powder under increasing quasistationary UV excitation. For an excitation power of about 2MW/cm^2 a laser mode at 2.23 eV reaches its threshold. The emission is localized to an area of about 2μm diameter (position A in the inset). At higher excitation powers more laser modes reach their threshold. Their emission originates from the whole area of the sample. The inset shows colour-coded luminescence intensity maps with spectral resolution on the abscissa and spatial resolution (intensity distribution along one narrow stripe of the ensemble) on the ordinate.

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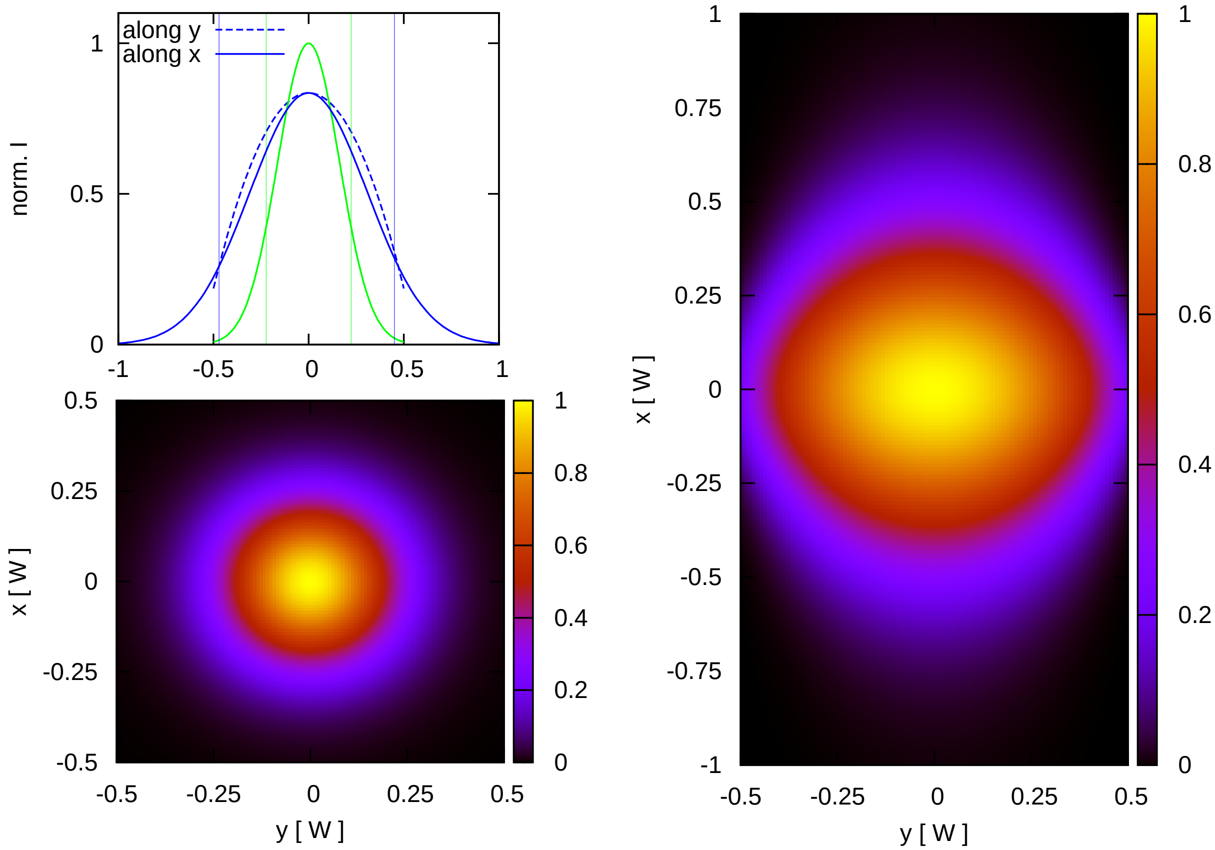

FIG. 2. Calculated mode diameters for a thin ($\sim 4.0\mu$m) ZnO random laser sample. The samples filling fraction is 50%, i.e. definitely in the strong scattering regime. The calculations are presented for the pump wavelength of $\lambda = 355nm$. The Mie scatterer's radii are assumed to be $r_0 = 0.36\lambda$. The finite sample width is estimated to be $W = 80r_0$, and for the calculation we consider the remaining spatial direction X to be infinitively extended. Numerically computed corresponding laser thresholds can be found in Fig. 3. The sample is here uniformly pumped. *Lower left panel:* Calculated confined mode. The pump intensity equals the experimental threshold intensity of the confined mode of $\sim 2.6MW/\mathrm{cm}^2$. The character of the circularly shaped confined mode can be clearly distinguished from the extended mode even though derived by the same formalism. Confined modes suffer an overall smaller loss of intensity. This loss mechanisms being perpendicular to the surface of the sample in the calculations can as such be clearly distinguished from those of the extended modes. *Right panel:* Calculated extended mode in stationary state. The pump intensity equals the threshold intensity derived from the experiment of $\sim 3.5MW/\mathrm{cm}^2$. The decay of the coherently emitted photon number with respect to the distance from the center of the mode is strongly renormalized due to the inherent differential attributed to the loss at the samples' edges at $Y = \pm 0.5$. That loss is assumed to be uniform along the edges and symmetric. The mode covers the entire sample in y-direction and it is elliptically shaped. *Upper left panel:* Comparisson of the intensity development along cuts through the mode center for the symmetric confined mode (green line) and the extended mode (blue lines, taken along x- and y-direction, respectively). The intensities are normalized to the threshold intensity of the confined mode in it's center. The influence of the differing loss mechanisms along the two directions is clearly visible.

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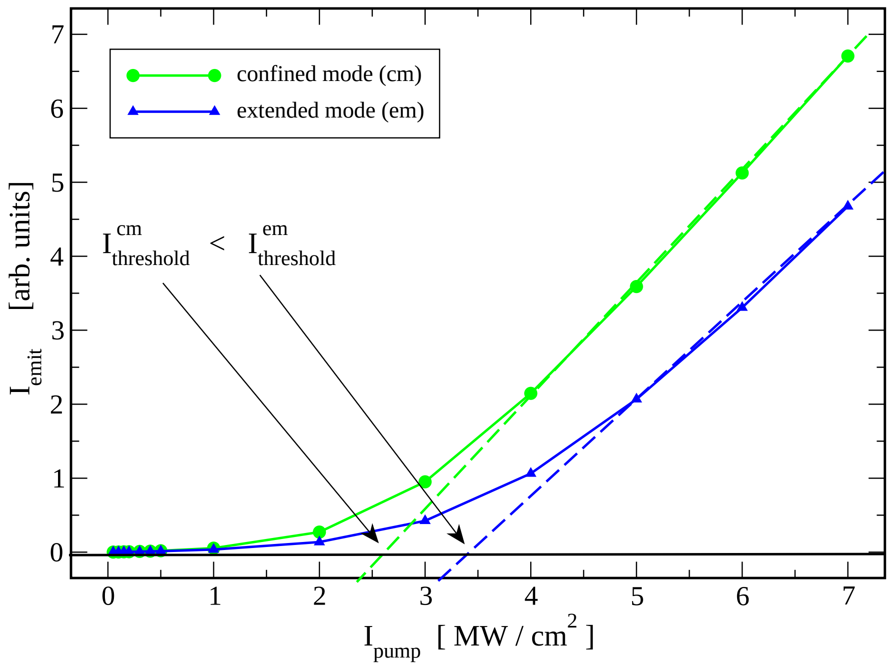

FIG. 3. Laser thresholds derived by the solution of the coupled system of mesoscopic transport and semi-classical time dependent laser rate equations (parameters can be found in caption of Fig. 2)). Extended and confined modes are suffering different types of loss by means of spontaneous emission and nonradiativ decay. The extended modes suffer in sum a stronger loss of intensity, and therefore arrive at their laser thresholds for significantly higher pump intensities. Due to the distinction of their loss mechanisms both types of modes do not compete with each other and therefore may spatially co-exist and overlap while they are spectrally well separated. Exactly this behavior has been found in our random laser experiments (see Fig. 1).

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Acknowledgments. First author (RF) thanks A. Lubatsch, F. Hasselbach, A. P. Mosk, G. Schön, K. Muttalib and P. Wölfle for highly valuable discussions. The authors acknowledge financial support by research grants of the Deutsche Forschungsgemeinschaft, KR 1726/3-1,-2 (RF), KL 354/23-1 + 2 (JS and HK). RF and HK are fellows of Karlsruhe School of Optics & Photonics (KSOP), all authors acknowledge KSOP for continuous support.